

# Cavity Enhanced Optical Vernier Spectroscopy

## Broad Band, High Resolution, High Sensitivity

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### Abstract

A femtosecond frequency comb provides a vast number of equidistantly spaced narrow band laser modes that can be simultaneously tuned and frequency calibrated with 15 digits accuracy. Our Vernier spectrometer utilizes all of these modes in a massively parallel manner to rapidly record both absorption and dispersion spectra with a sensitivity that is provided by a high finesse broad band optical resonator and a resolution that is only limited by the frequency comb line width while keeping the required setup simple.

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Femtosecond frequency combs [1] provide an equidistant array of narrow line width frequencies that can be simultaneously tuned. In this paper we demonstrate how to employ such a frequency comb to record broad band, high sensitivity absorption *and* dispersion spectra with potential Hertz resolution in very short time. The high sensitivity is provided by means of a simple Fabry Perot resonator that vastly extends the interaction length with the sample under study and has the potential to achieve sensitivities of classical ring down techniques. The Hertz resolution is provided by an appropriately stabilized laser [2]. As the frequency comb can readily be referenced to a primary frequency standard, the frequency accuracy and reproducibility can be as good as  $10^{-15}$ . Such a system could prove useful for quick and reliable identification of molecular species with complex amplitude and phase structure. To achieve comb mode resolution while keeping the setup simple, the resonator acts as a filter that thins out the frequency comb such that each individual mode can be resolved with the help of a small diffraction grating. The resonator modes are scanned across the frequency comb like a Vernier scale in frequency space. Groups of comb lines that contain the spectroscopic data are streaked on a two dimensional array. In this way a comparatively slow detector such as a charge coupled device (CCD) can be used to quickly acquire data. Similar to a previous method [3] it allows to determine the dispersive details of the resonator itself which is essential for designing femtosecond enhancement resonators that have recently been used to generate frequency combs in the extreme ultraviolet and beyond [4, 5].

In the last decade a variety of methods were introduced, which exploit the unique combination of broad bandwidth and high temporal coherence of a frequency comb for sensing and identifying (“fingerprinting”) chemical species [3, 6, 7, 8, 9, 10, 11]. Dispersion compensated Fabry Perot resonators exhibit an equidistant grid of resonances that can be lined up with the modes of the frequency comb, which has the consequence that the entire driving power can be simultaneously resonant. Cavity enhanced absorption spectroscopy methods (like cavity ring down) take advantage of this fact to achieve a higher signal to noise ratio [8, 9, 10, 11]. The resolution of these methods was however limited by the spectrometer dispersing the light after its passage through the resonator. Resolving individual comb components on the other hand can boost the resolution down to the Hertz level that can only be provided by stabilized lasers [2]. Doing so over a large bandwidth with a grating proves challenging as such a grating has to be very large and expensive if operated in first order. If

operated in higher order the bandwidth is limited due to overlapping diffraction orders. An effective and simple method to simultaneously resolve and record a vast number of individual comb modes has recently been demonstrated [12]. It combines two spectrally dispersing elements in an arrangement similar to a prism-echelle spectrometer. One of these operates in high order and provides comb resolution, while the second operates in first order to separate the overlapping diffraction orders of the first. This approach is however difficult to combine with a broadband ring down technique [10] as both methods require a two dimensional array for detection.

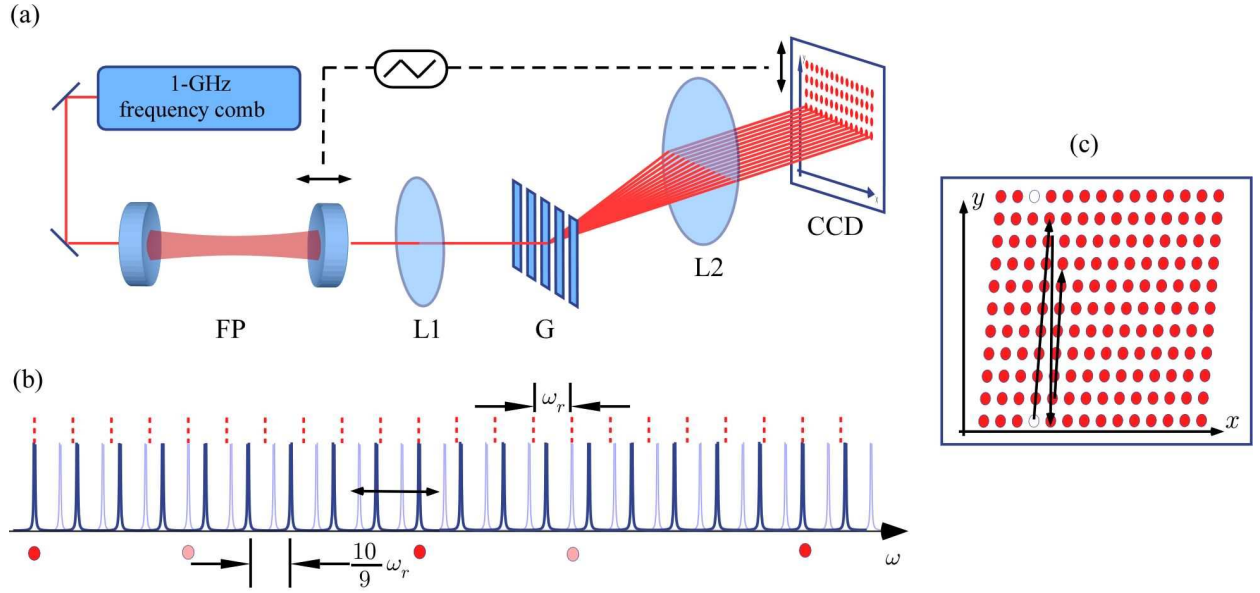


FIG. 1: a) General setup: A femtosecond laser output is sent to a Fabry Perot (FP) resonator with a scanning mirror. The transmitted light is spectrally resolved by a grating spectrograph. Synchronously with the FP, the CCD is scanned orthogonal to the spectrograph dispersion to map the transmission spectrum of the FP as a function of cavity tuning to the  $y$  axis on the CCD. L1,L2: lenses, G: grating. Panel b) frequency comb (dashed) transmission function of the resonator (solid). The resonator is detuned to the comb to provide a Vernier for the frequency comb, which is shifted as the resonator length changes. Dots mark transmitted modes for two positions of the Vernier. Panel c) Expected pattern on the screen behind the spectrometer. Arrows mark the order of the frequency comb modes (index increasing by one from spot to spot along the arrows, open spots belong to the same comb frequency)

In our optical Vernier spectrometer (fig. 1), this difficulty is circumvented by combining

the comb mode resolving, high diffraction order part of the echelle spectrometer [12] and the sensitivity enhancing high finesse resonator into one device. The output of a frequency comb laser has a spectrum containing only an equidistant grid of frequencies  $\omega_k = \omega_{CE} + k\omega_r$ , where  $\omega_r$  is the pulse repetition frequency of the laser,  $k$  is a large integer and the components are offset from being integer multiples of  $\omega_r$  by the carrier envelope offset frequency  $\omega_{CE}$ . This light is sent to the high finesse resonator, which contains the sample under study. It has resonances spaced by its free spectral range  $FSR = 2\pi c/nL$ , where  $c$  is the speed of light in vacuum and  $n$  is the effective refractive index. The geometrical length  $L$  of the resonator is adjusted such that, if the two scales are lined up, every  $m$ -th mode from the frequency comb will be resonant with every  $n$ -th mode of the cavity and transmitted through it. Provided the finesse of the resonator is high enough (i.e. larger than  $m$ ), the other modes will be strongly suppressed. In this way the modes of the frequency comb and the resonant frequencies of the resonator resemble a Vernier (fig. 1b) with the *Vernier ratio*  $FSR/f_r = m/n$  being a cancelled fraction. In our case this is chosen to be  $69/68$  ( $10/9$  in the figure for clarity). By choosing  $m = n + 1$  the interaction length in the resonator is essentially the same as in a matched resonator, instead of  $1/m$  for  $n = 1$  which would significantly reduce the sensitivity. Then the frequency comb that is transmitted through the resonator has a spacing of about  $m \times 1$  GHz, so that for sufficiently large  $m$  it can easily be resolved with a very simple and small grating spectrograph. As the length of the resonator is now tuned, the Vernier primarily shifts, bringing the next set of  $m \times 1$  GHz spaced comb modes into resonance. After scanning the resonator length by half a wavelength (i.e. one free spectral range),  $m$  groups of  $m \times 1$  GHz combs have passed by and the situation is as in the beginning, the initial group is transmitted. The only difference is that the Vernier ratio has slightly changed due to the change in resonator length.

Now a mirror rotates synchronized with the scanning resonator to streak the light transmitted through the resonator along the axis (y-direction) that is perpendicular to the grating dispersion (x-direction). A 2-dimensional CCD detector is (also synchronized to the scanning) exposed to the output of that spectrometer (in figure 1a a moving detector is shown for simplicity). This results in a 2-dimensional array of spots on the CCD that can uniquely be ordered into a single frequency axis as shown in fig. 1c, very similar to the sorting procedure used in ref. [12]. Like this we can associate the comb mode frequency  $\omega_{i,j} = \omega_{k0+i+mj}$  to the spot in row  $i$  and column  $j$  (counting from an arbitrary spot  $(0,0)$ , belonging to frequency

$\omega_{k0}$ ). Both the precise value of  $m$  and  $k0$ , and therefore absolute frequency calibration of the data can easily be determined if there is a sufficiently narrow feature of known (better than  $f_r$ ) frequency in the spectrum (e.g. an absorption resonance, blue spots in fig. 1c). This feature will repeat itself as the resonator is tuned and the period (in spots) gives the correct value for  $m$ . Of course because of the spectrum is sampled at comb frequencies only, the resolution of a single such pattern is on the order of  $f_r$  and aliasing effects may occur. An essentially arbitrarily high resolution can be achieved by recording a series of spectra with different center frequencies of the frequency comb to fill the gaps.

The signal of the spectroscopy is contained in the brightness and shape of the spots as well as their position. To quantitatively interpret this pattern, we consider a simple steady state model for the signal that is observed on the CCD as a function of frequency (x-axis on the CCD) and resonator length (y-axis on the CCD). Assuming the individual comb modes can be well isolated from each other on the CCD image, we can approximate the transmission function in the vicinity of spot  $(i, j)$  as a simple Airy function originating from a single comb mode  $\omega_{i,j}$ , i.e.

$$\mathcal{T}_{i,j}(x, y) = \frac{T^2 \delta(\omega(x) - \omega_{i,j})}{1 + r(\omega_{i,j})^2 - 2r(\omega_{i,j}) \cos(\phi(L(y), \omega_{i,j}))}, \quad (1)$$

where  $r$  is the round trip amplitude loss factor  $r(\omega) = \sqrt{1 - A(\omega)}(1 - T)$ ,  $T$  the resonator mirror transmission,  $A(\omega)$  the absorption of the medium inside the resonator per round trip,  $\delta$  the line shape function of the individual frequency comb mode and  $\omega(x)$  and  $L(y)$  are appropriate calibration functions of the spectrometer. The line shape function  $\delta$  may be approximated by a Dirac delta function for the most practical purposes. The round trip phase shift  $\phi(L, \omega) = \psi(\omega, L) + L\omega/c$  contains a vacuum contribution  $L\omega/c$  and the additional phase shift  $\psi(\omega, L)$  due to dispersion of the mirrors and the medium. This Airy function (1) assumes an infinitely large aperture spectrometer and a point source and hence an infinite resolution. With the parameters chosen in this demonstration experiment however, the actual spot shapes are dominated by the laser beam profile, which can be approximated by a 2-dimensional Gaussian of fixed width. The convolution of (1) with this beam profile may be fit to each spot  $(i, j)$  in the image to obtain both  $A(\omega_{i,j})$  and  $\psi(\omega_{i,j})$  up to multiples of  $2\pi$ , provided  $y$  can be calibrated absolutely to the resonator length  $L$  ( $\omega(x)$  need not be known). We derived the scaling for  $L(y)$  from the fact that  $\omega_{i,j}$  and  $\omega_{i+m,j-1}$  represent the same frequency, so that the phase shift difference between the two spots needs

to be  $2\pi$  and therefore  $L(y_{i+m,j-1}) - L(y_{i,j}) = 2\pi c/\omega_{i,j}$ . Here  $y_{i,j}$  represents the y-position of the maximum of the peak on the CCD. The dependence of  $\psi(L, \omega)$  on the resonator length was neglected in this expression. This is a good assumption as long as the extended medium has a refractive index very close to unity (or has a fixed length). Using this relative length calibration, one may determine  $\psi(\omega)$  up to a linear function.

We use a 1 GHz mode locked Ti:Sapphire laser with a bandwidth of about 12 THz FWHM centered at 785 nm and an average output power of about 0.5 W as a light source [13]. The spectroscopy resonator consists of broadband dielectric quarter wave stack mirrors, centered at 792 nm and arranged in a bowtie ring cavity configuration. The FSR is about  $(69/68)f_r$ . The two coupling mirrors dominate the resonator loss with their nominal transmission of 0.1%, corresponding to a resonator finesse of  $1000\pi$ , which we confirmed by ring down measurements with the laser operated in a continuous mode. One of the coupling mirrors is mounted on a piezo electric transducer tube for scanning the resonator length. The buildup time of  $1 \mu\text{s}$  sets a lower limit of about 3 ms on the scan time over a full free spectral range in order to satisfy the steady state approximation that was used in (1). We use a cm size 2100 lines per mm holographic grating in a 150 mm focal length Cherny-Turner arrangement. The aperture of the grating is fully used to match the resolution of the spectrometer to the pixel size ( $5 \mu\text{m}$ ) of  $1000 \times 1200$  pixel CCD camera that was used to record the images (WinCAM-D 1M4). The resolution of the spectrometer expected from these parameters is better than 30 GHz at 760 nm. Raw data in the wavelength range from 760 to 770 nm with air inside the resonator were obtained in the way described above and are shown in figure 2. This image was exposed for 9 ms, the time for one scan.

After acquisition, the period of an observed absorption pattern was used to confirm the detuning ratio of the resonator to be 69/68. To obtain brightness values for each spot, a 2-d Gaussian was fit to the the spot and the its integral was used as a brightness value. This is a good approximation to the model (1), as explained above. Because this is the case, the integral was related to the Airy (1) area to obtain an intracavity absorption value. Figure 3a shows an absorption spectrum obtained like this after applying a linear filter to the data (see below). A comparison with the HITRAN [14] molecular spectroscopic database reveals that the absorption feature is caused by a  $X^3\Sigma_g^- \rightarrow b^1\Sigma_g^+$  magnetic dipole intercombination transition in molecular oxygen [15]. We used this identification to calibrate the offset frequency  $\omega_{CE}$ , that was not measured independently in this demonstration. Note

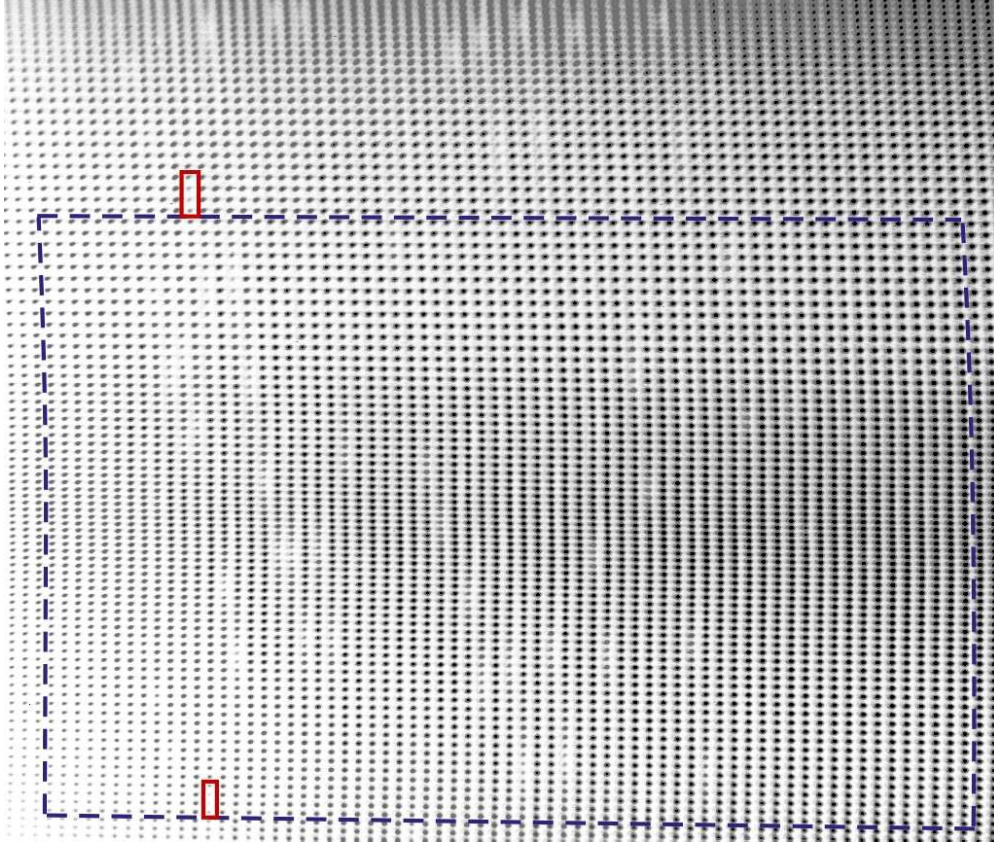


FIG. 2: Raw data image as seen by the CCD, white corresponds to no light. The horizontal axis is the dispersion axis of the grating and the vertical axis is the scan axis. Dark spots caused by absorption lines in the A-bands of  $O_2$  are clearly observed. The period in the pattern in vertical direction identifies the detuning to 69/68th (solid boxes mark exemplary one pattern that was identified). The dashed box marks a unique data set. The varying spot spacing and brightness common to all spot columns originates from a nonlinearity of the scan and is corrected for in the data analysis.

that our measurement perfectly agrees in frequency scale, amplitude and line width with the database values (fig. 3).

One strength of the method is to provide both amplitude and phase information about the sample. The phase information that could be extracted is shown in figure 3. This (round trip) phase spectrum has a suitable linear function subtracted in order to make the dispersion more easily be seen. It shows a global second order dispersion, as expected from the dielectric mirrors that were used at the blue end of the mirrors' reflectance. On top of that local dispersive features are seen, which agree, in position and amplitude, with the expected



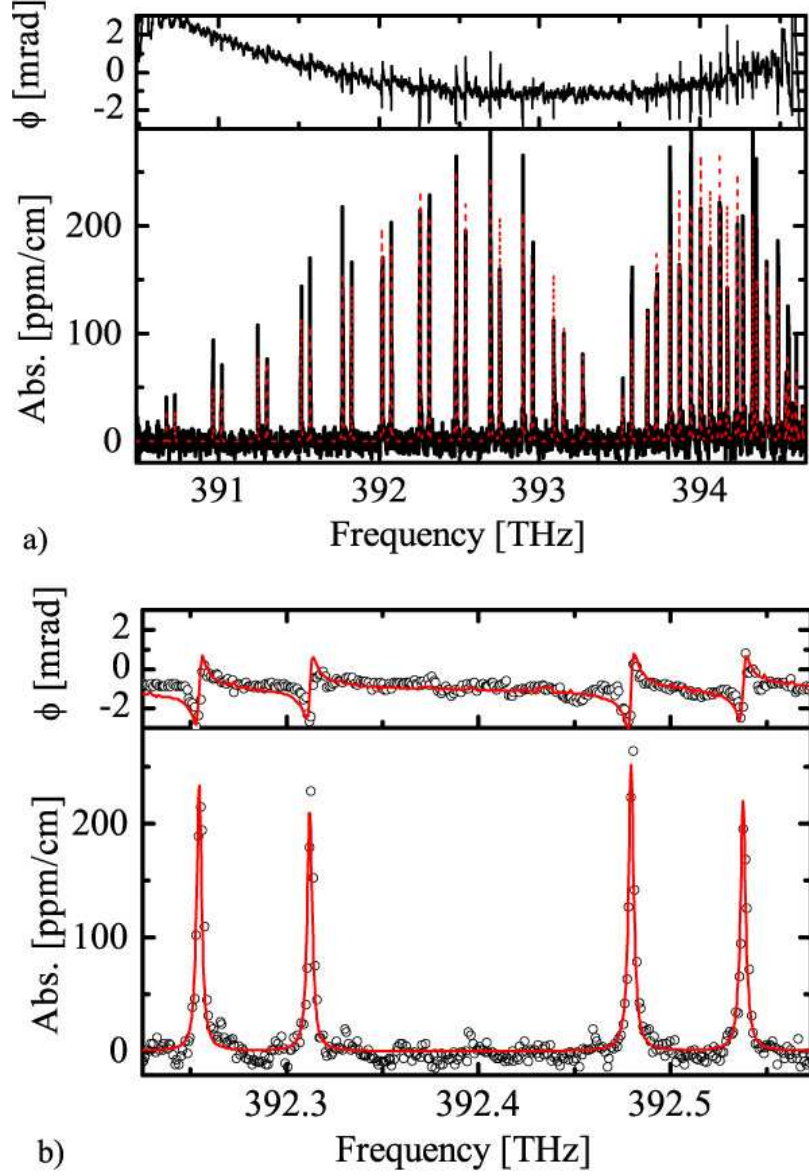


FIG. 3: Panel a) shows the entire spectrum extracted from the raw data shown in fig. 2 (solid line), which is in good agreement with the HITRAN database values [14] (dashed red line). The phase shows a global positive second order dispersion of the resonator as compatible with the mirror design characteristics and spikes at the resonances. The only free parameter, a global frequency offset, we obtained by matching one absorption peak to the HITRAN data. Panel b) shows a zoom of the dataset, highlighting the excellent agreement between our data (circles) and the HITRAN database (red line).



dispersive features of the O<sub>2</sub> resonances. The phase sensitivity with the current implementation is not specifically high, as the Airy pattern is not resolved by the spectrometer so that the phase information is degraded.

By evaluating the rms absorption noise level at positions where no signal is detected, we estimate the absorption sensitivity of the method for a single frequency to be  $5 \times 10^{-6}/\text{cm}$  for a single image taken within 10 ms. With an appropriate camera with no (small) dead times between acquisitions, this can lead to a sensitivity of  $5 \times 10^{-7}/\text{cm}\sqrt{\text{Hz}}$  already for this proof-of-principle experiment. Additionally, the shown spectrum was recorded in the far wing of the laser spectrum where little power was available and one can expect the signal to noise to increase with more driving power.

The most severe limitation in this preliminary implementation comes from acoustic noise and the piezo transducer's imperfections, which render the length scan of the resonator mirror nonlinear, leading to varying vertical positions and brightness of the observed spot pattern. However, the scan position is the same for any spot with the same y-value and such distortions will be the same for every spot column. The higher spot density in the vertical center of the data fig. 2 is an example of such a pattern. An appropriate linear filter was used to reduce the magnitude of this artifact and the result is shown in fig. 3. Of course, this procedure will also reject real structure that by accident has the same property. Such a case can be tested for by changing the resonator detuning, which in turn will make the real structure reappear. Still this and other noise sources reduce the sensitivity from the shot-noise limited  $3 \times 10^{-9}/\text{cm}$  to the observed level of  $5 \times 10^{-6}/\text{cm}$  in 10 ms, and does particularly also degrade the quality of the phase data. In an improved version, this excess noise can be eliminated by using a stable optical reference (diode laser, offset locked to the frequency comb) to which the resonator can be locked. With this and resolved Airy fringes, it should be possible to considerably reduce the excess noise on both amplitude and phase outputs.

In conclusion, we have presented a simple method that provides comb line resolution over a band width of more than 4 THz, which should be easily extendible to 40 THz by packing the spot pattern more tightly. Simultaneously the sensitivity is better than  $10^{-6}/\text{cm}\sqrt{\text{Hz}}$  per mode which was achieved in an interaction length of 29 cm only using a moderately high finesse optical resonator. The method can be made equivalent to a ring down method by increasing the scan speed and replacing (1) with an appropriate non steady state version [16]

in order to get into a real ring down regime. Then, if the streak speed is chosen appropriately so that the ring down wave form can be seen, the decay time may be separated from the influence of technical noise. In addition to ring down methods it also provides dispersion information with a potential accuracy equivalent or exceeding the one presented in [3]. Due to the scanning resonator approach, it is not necessary to accurately control intracavity dispersion, which in other methods results in a reduced transmission or even spurious signals. The price that has to be paid for these advantages compared to matched resonators is, that the average power transmission through the resonator is  $1/\text{finesse}$  so that the shot noise limited absorption sensitivity only scales as the square root of the finesse instead of the finesse itself. Yet our method is fast. A single acquisition takes only about 10 ms giving 4000 data points (4 THz with a resolution of 1 GHz). With little optimization, one can easily obtain 20-40 thousand spots on a mega pixel CCD (the only requirement being that the spots are well separated). The resolution of the scheme is essentially unlimited (Hz level, due to potential laser line width). And it can, at any time, be referenced to a primary frequency standard, giving a frequency reproducibility and accuracy of  $10^{-15}$  (sub-Hertz level!), which is limited by the primary standard only. The potentially high resolution naturally suggests the application of the demonstrated technique to some kind of cavity enhanced nonlinear Doppler free spectroscopy [17, 18].

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